

# Stability of a Thin Solid Film with Interactions

Vijay Shenoy<sup>†</sup> and Ashutosh Sharma<sup>‡</sup>

Indian Institute of Technology Kanpur, UP 208 016, India

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We investigate the question of stability of a solid thin film which experiences external interactions such as van der Waals forces from a contacting surface or forces from an external electric field. Both perfectly elastic and viscoelastic material behaviours are considered in linear stability analysis performed here. These analyses indicate that for sufficiently soft (shear modulus between 1 and 10 MPa) and nearly incompressible films (Poisson's ratio close to 0.5), bifurcations are possible, i. e., the surface of the film becomes non-planar. The modes of bifurcation and rates of growth of perturbations are determined as a function of material parameters. The results of this study are of significance in understanding the adhesive properties between a soft material (such as rubber) and a comparatively rigid solid (such as steel), and the behaviour of soft solid films in an electric field.

Instabilities and pattern formation in thin solid and liquid films are of interest both from a scientific and technological view point. Morphological instabilities in thin liquid films occur due to causes such as competition between capillary forces and van der Waals interactions [1] or an external electric field [2] and can often lead to dewetting leading to interesting patterns. Morphological instabilities are also common in solid films; for example, in stressed solid films the strain energy drives the surface roughening in competition with the surface energy with surface mass diffusion being the dissipative mechanism[3, 4].

Analysis of interacting thin films has hitherto been restricted to fluid films. Here, we pose the question of stability of a thin *solid* film bonded to a rigid substrate whose free surface experiences an effective force. This force may arise from any of the various causes such as a van der Waals interaction with another contacting surface nearby and/or with the substrate, an external electric field, etc. The theoretical analysis presented in this paper indicates that for a *soft and nearly incompressible* solid thin film, instabilities are possible and that the film “buckles”. Physically, this instability occurs because it is possible, for sufficiently large interaction forces, to reduce the net potential energy of the system (the elastic strain energy and the surface energy of the film + potential energy of interaction of the surface) by a periodic non homogeneous deformation in the film. We believe that these results could be useful in understanding phenomena of adhesion between materials (such as rubber and steel), behaviour of thin films in an electric field etc.

The system considered here is shown in fig. 1 – a film of height  $h$  bonded to a rigid substrate described by coordinates  $(x_1, x_2)$  such that surface of the film  $S$  interacting with external agency has  $x_2 = 0$  and that bonded with the rigid substrate has  $x_2 = -h$ . We restrict attention to plane strain deformations of

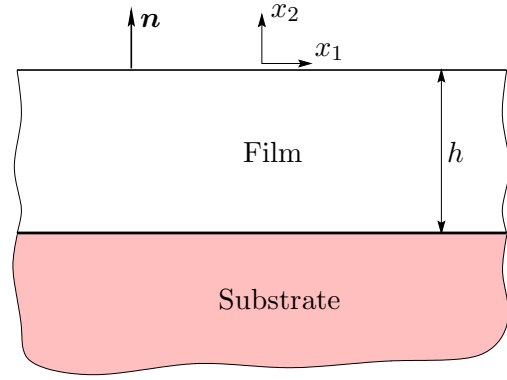


FIG. 1: Film bonded to a rigid substrate. The surface of the film experiences external forces.

the film for the sake of mathematical simplicity and to understand the essential physics. The total potential energy of this system is

$$\int_V W(\epsilon) dV + \int_S \left( \gamma \sqrt{1 + (u_{2,1})^2} - U(\mathbf{u} \cdot \mathbf{n}) \right) dS \quad (1)$$

where  $\epsilon$  is the strain tensor,  $W(\epsilon)$  is the elastic strain energy density,  $\gamma$  is the surface energy,  $U(\mathbf{u} \cdot \mathbf{n})$  is the interaction potential between the surface of the film and the external agency such as a contactor or an electric field,  $\mathbf{u}$  is the displacement vector and  $\mathbf{n}$  is the outward normal to the surface. Linearised analysis is performed by expanding the interaction term  $U(\mathbf{u} \cdot \mathbf{n})$  in a power series about  $\mathbf{u} = \mathbf{0}$  and retaining all terms up to quadratic order in  $\mathbf{u}$ . The resulting approximate energy functional is

$$\int_V W(\epsilon) dV + \int_S \gamma \sqrt{1 + (u_{2,1})^2} dS - \int_S \left( U_0 + F_0 \mathbf{u} \cdot \mathbf{n} + \frac{1}{2} Y (\mathbf{u} \cdot \mathbf{n})^2 \right) dS \quad (2)$$

where

$$U_0 = U(0), \quad F_0 = U'(0) \quad \text{and} \quad Y = U''(0). \quad (3)$$

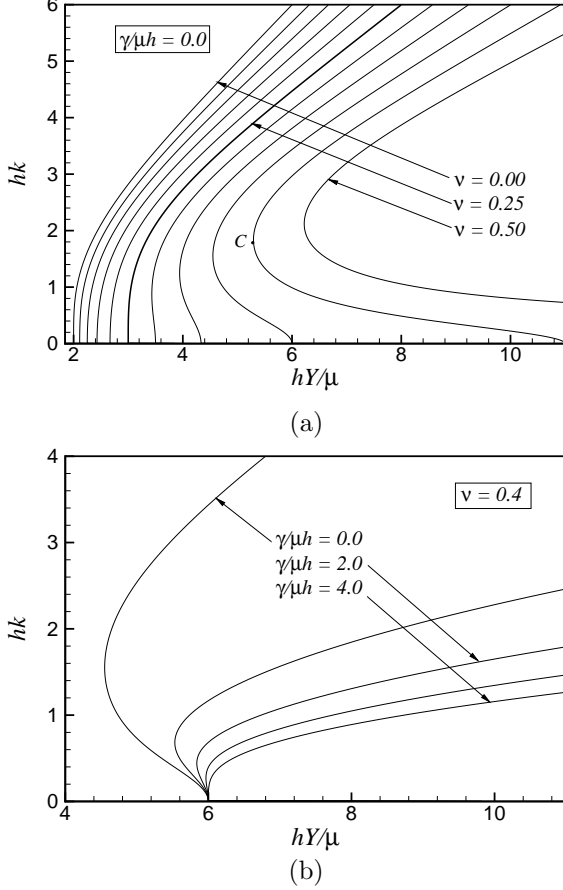


FIG. 2: Bifurcations modes ( $hk$ ) as a function of  $hY/\mu$  for various values of  $\nu$  with  $\gamma/\mu h = 0$ . (b) Bifurcations modes ( $hk$ ) as a function of  $hY/\mu$  for various values of  $\gamma/\mu h$  with  $\nu = 0.4$ .

The equilibrium stress field  $\boldsymbol{\sigma}$  in the film (which minimises the potential energy (2) over an appropriate length of the film) satisfies the equilibrium equation  $\nabla \cdot \boldsymbol{\sigma} = \mathbf{0}$  in  $V$  and the boundary condition

$$\boldsymbol{\sigma} \cdot \mathbf{n} = \gamma u_{2,11} \mathbf{n} + F_o \mathbf{n} + Y(\mathbf{u} \cdot \mathbf{n}) \mathbf{n} \quad (4)$$

on  $S$ . Taking the film to be an isotropic linear elastic solid with shear modulus  $\mu$  and Poisson's ratio  $\nu$ , gives a standard expression for the strain energy density [5] with a resulting expression for the stress tensor expressed in terms of the gradient of displacement. Thus the problem can be cast into a boundary value problem for the unknown displacement field with the boundary condition of vanishing displacements at  $x_2 = -h$  at the film substrate interface in addition to (4).

*The Homogeneous Solution:* A solution to the above boundary value problem exists such that the stresses in the film are equal everywhere. This homogeneous solution ( $\mathbf{u}^h$ ) is  $u_1^h = 0$  everywhere, and  $u_2^h$  has a linear variation with  $x_2$  starting from 0 at  $x_2 = -h$ , i.e.,

$$u_2^h(x_1, 0) = u_o = \frac{F_o}{\left(\frac{2(1-\nu)\mu}{(1-2\nu)h} - Y\right)}. \quad (5)$$

For the case when  $\nu = 0.5$ , i. e., the incompressible

limit, the homogeneous solution is such that the displacement vanishes everywhere in the film, and a pressure field  $p$  develops such that  $p(x_1, x_2) = F_o$ . So long as

$$Y < Y_m, \quad \frac{hY_m}{\mu} = \frac{2(1-\nu)}{(1-2\nu)} \quad (6)$$

the homogeneous solution is meaningful in that  $u_o$  has the same sign as  $F_o$ . This conditions on  $Y$  is most easily met when  $\nu$  is close to 0.5 (the r.h.s. of (6) tends to  $\infty$  as  $\nu$  tends to 0.5), i. e., when the material is nearly incompressible. It is this class of materials that the focus of this paper. Nevertheless, results are presented for all values of  $\nu$  for the sake of completeness.

*Bifurcations:* What are the conditions (on  $Y, \mu, \nu, h, \gamma$ ) for another solution (inhomogeneous state) to exist? If such a solution exists, it can be taken to be of the form  $\mathbf{u}^h + \mathbf{u}$ , where the symbol  $\mathbf{u}$  now stands for a ‘‘bifurcation’’ displacement field. This bifurcation field must satisfy the equilibrium equations in the bulk and the rigid boundary condition at the film substrate interface, just as the homogeneous solution. On the surface of the film at  $x_2 = 0$ , the bifurcation field satisfies (here  $\boldsymbol{\sigma}$  is the *additional* stresses due to  $\mathbf{u}$ ),

$$\boldsymbol{\sigma} \cdot \mathbf{n} = \gamma u_{2,11} \mathbf{n} + Y(\mathbf{u} \cdot \mathbf{n}) \mathbf{n}, \quad (7)$$

instead of (4). To investigate the existence of a nontrivial solution to the problem defined above, the bifurcation fields are assumed to have the form

$$u_j(x_1, x_2) = e^{ikx_1} u_j(x_2) \quad (8)$$

where  $k$  is a real positive wavenumber. The problem of finding nontrivial bifurcation fields can be cast into the problem of finding those values of  $k$  such that the functions  $u_j(x_2)$  are nontrivial. It can be shown that (a detailed account will be published elsewhere) nontrivial bifurcation fields of the form (8) exist for those values of  $k$  that satisfy the equation

$$\left( k \left[ 4e^{2hk} h k^2 (h\mu - (1-\nu)\gamma) + (e^{4hk} - 1) k \gamma (3 - 7\nu + 4\nu^2) + \mu \left( (3 - 4\mu)(1 + e^{4hk}) - 2e^{2hk} (5 - 12\nu + 8\nu^2) \right) \right] \right) / \left( (1-\nu) \left[ (3 - 4\nu)(e^{4hk} - 1) - 4hk e^{2hk} \right] \right) = Y \quad (9)$$

This relation is valid for the incompressible case as well (i. e., when  $\nu = 0.5$ ). Real roots of (9) are sought when  $Y < Y_m$  which is the range of  $Y$  for which the homogeneous solution is valid.

We first focus attention on the case when  $\gamma$  vanishes. Fig. 2a depicts graphically the solution to (9), i. e., for a given value of  $\nu$ , the values of  $k$  that solve (9) are plotted as a function of  $Y$  ( $hY/\mu$  in non-dimensional terms). The important results may be noted: (i) There are no bifurcation modes for any value of  $\nu$  when  $hY/\mu < 2$ . (ii) For all values of  $\nu$ ,  $k = 0$  is a bifurcation mode when  $Y = Y_m$ . (iii) When  $\nu \leq 0.25$ , there are no bifurcation modes for  $Y < Y_m$ . (iv) When  $\nu > 0.25$ , there are two modes starting from a critical

value  $Y_c$  (such as the point  $C$  shown in fig. 2a) that depends on the value of  $\nu$  until  $Y$  reaches  $Y_m$ . When the film is incompressible  $hY_c/\mu = 6.22$  and the corresponding bifurcation mode has  $hk_c = 2.12$ . For this case bifurcations are possible for all values of  $Y$  greater than  $6.22\mu/h$ , with two possible values of  $k$  as shown in the fig. 2a.

Next, we consider the case when  $\gamma \neq 0$ . Fig. 2b shows a plot of the possible wavenumbers of bifurcation modes for various values of  $\gamma$  with  $\nu = 0.4$ . The key effect of the surface energy on the bifurcation modes are noted as follows: (i) Surface energy inhibits bifurcation, in that a larger value of  $Y_c$  is effected with a non zero value of  $\gamma$ . The critical mode  $k_c$  decreases with increasing  $\gamma$ . Both of these results are as expected since a larger value of  $k$  implies a larger energy penalty in terms of surface energy. (ii) As  $\gamma$  gets larger  $Y_c$  approaches  $Y_m$ . In fact, it can be shown that  $Y_c$  equals  $Y_m$  when  $\gamma = \gamma_m$  where

$$\frac{\gamma_m}{\mu h} = \frac{2\nu(4\nu - 1)}{3(1 - 2\nu)^2}, \quad (10)$$

a result which is pertinent when  $\nu > 0.25$ . The curve for  $\gamma/\mu h = 4.0$  for the case of  $\nu = 0.4$  shown in fig. 2, graphically illustrates this point. If  $\gamma > \gamma_m$ , then there are no bifurcations in the physically meaningful range  $Y < Y_m$ .

A more detailed analysis gives the following formulae for  $Y_c$  and  $k_c$  as a function of  $\gamma$  and  $\nu$  when  $\gamma/\mu h \ll 1$  and  $\nu \rightarrow 0.5$ :

$$\begin{aligned} \frac{h}{\mu} Y_c(\nu, \gamma/\mu h) &= 6.22 - 10.46(1 - 2\nu) + 4.49 \frac{\gamma}{\mu h}, \\ hk_c(\nu, \gamma/\mu h) &= 2.12 - 2.86(1 - 2\nu) - 2.42 \frac{\gamma}{\mu h}. \end{aligned} \quad (11)$$

It is also interesting to consider the time evolution of deformation in the film so as to obtain the dominant or the fastest growing mode. To this end, the film is considered to be viscoelastic with a constitutive relation of the form

$$\begin{aligned} \boldsymbol{\sigma} &= 2\mu \left( \frac{1}{2}(\nabla \mathbf{u} + \nabla \mathbf{u}^T) + \frac{\nu}{1 - 2\nu} \nabla \cdot \mathbf{u} \mathbf{I} \right) \\ &+ 2\eta \left( \frac{1}{2}(\nabla \dot{\mathbf{u}} + \nabla \dot{\mathbf{u}}^T) - \frac{1}{3} \nabla \cdot \dot{\mathbf{u}} \mathbf{I} \right), \end{aligned} \quad (12)$$

where  $(\dot{\phantom{x}})$  stands for the time derivative,  $\eta$  is a viscosity parameter and  $\mathbf{I}$  is the second order identity tensor. In the consideration of the time evolution of the system, inertial effects are neglected since the time scale of interest is much larger than the time scale of the propagation of an elastic wave through the thickness of the film.

*The Homogeneous Viscoelastic Solution:* The homogeneous solution of the field equations with the viscoelastic constitutive relation (12) is

$$u_1^h = 0, \quad u_2^h(x_1, x_2, t) = u_0 \left( 1 + \frac{x_2}{h} \right) \left( 1 - e^{\omega^h t} \right) \quad (13)$$

where  $\omega^h$  is given by

$$\omega^h = -\frac{3}{4\eta} \left( \frac{2(1 - \nu)\mu}{(1 - 2\nu)h} - Y \right) = -\frac{3}{4\eta} (Y_m - Y). \quad (14)$$

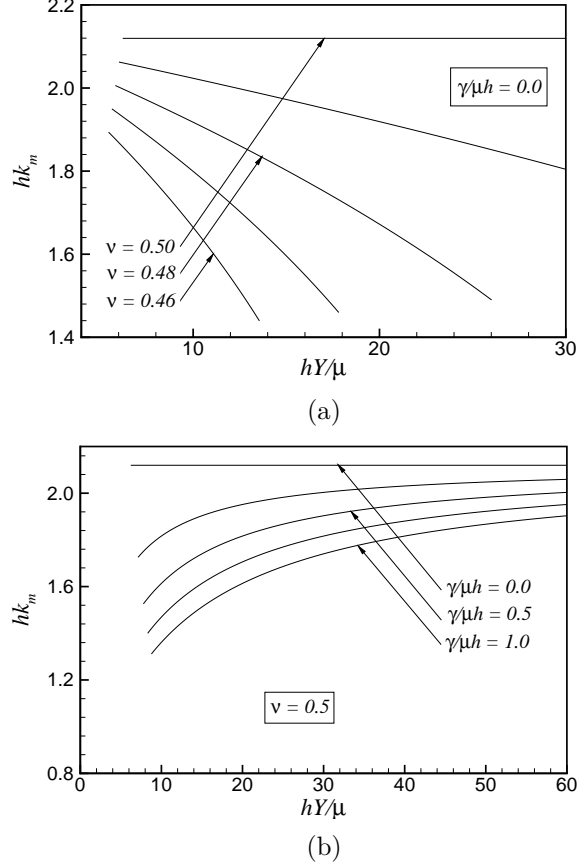


FIG. 3: Bifurcations modes ( $hk$ ) as a function of  $hY/\mu$  for various values of  $\nu$  with  $\gamma/\mu h = 0$ . (b) Bifurcations modes ( $hk$ ) as a function of  $hY/\mu$  for various values of  $\gamma/\mu h$  with  $\nu = 0.4$ .

From (14) it is evident that the time dependent homogeneous solution tends to the elastic homogeneous solution as  $t \rightarrow \infty$  when  $Y < Y_m$ . If  $Y > Y_m$ , the present analysis indicates that the homogeneous solution blows up as  $t \rightarrow \infty$ .

*Growth of Perturbations:* Just as in the case of the elastic film, it is of interest to investigate the growth of perturbations of the homogeneous solution. The perturbations  $\mathbf{u}$  are assumed to be of the form

$$u_j(x_1, x_2, t) = e^{ikx_1} u_j(x_2) e^{\omega t}. \quad (15)$$

For a given  $k$ , the rate of growth  $\omega$  is determined by insisting that the the perturbation satisfies equilibrium equations and boundary conditions and that they be nontrivial. The relation between  $\omega$  and  $k$  can be obtained by replacing  $\mu$  and  $\nu$  in (9) respectively by  $\mu^*$  and  $\nu^*$  where,

$$\mu^* = \mu + \eta\omega, \quad \nu^* = \frac{3\nu\mu - (1 - 2\nu)\eta\omega}{3\mu + (1 - 2\nu)\eta\omega}. \quad (16)$$

This procedure results in a cubic equation for  $\omega$ . The solution of this equation is obtained by numerical means.

The solution for  $\omega$  indicates that for  $Y_c < Y < Y_m$ , all perturbation modes with wavenumbers between the two bifurcation modes given by the elastic analysis

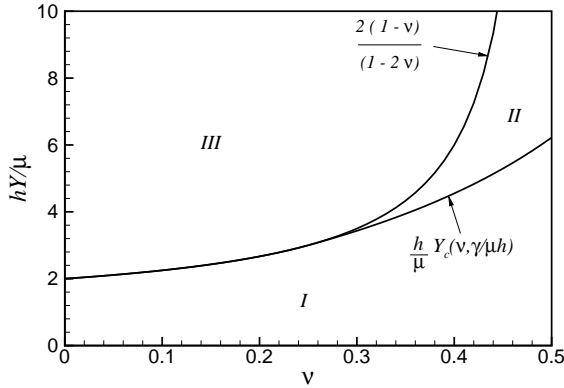


FIG. 4: The stable and unstable regions in the parameter space. Region *I*: Homogeneous solution stable. Region *II*: Homogeneous solution unstable with two elastic bifurcation modes and a fastest growing viscoelastic perturbation. Region *III*: Homogeneous solution “unphysical”.

are unstable i. e.,  $\omega$  for these modes are positive. Indeed, there is a mode with wavenumber ( $k_m$ ) between wavenumbers of the two elastic bifurcation modes such that the rate of growth ( $\omega$ ) is a maximum. Fig. 3(a) shows a plot of  $k_m$  as a function of  $Y$  ( $Y_c \leq Y \leq Y_m$ ) for various values of  $\nu$  (with  $\gamma/\mu h = 0$ ). When  $\nu < 0.5$ , the value of  $k_m$  starts at  $k_c$  when  $Y = Y_c$  and monotonically falls with increasing  $Y$ . For the case of  $\nu = 0.5$ ,  $k_m = k_c$  for all values of  $Y$ . When  $\gamma \neq 0$ ,  $k_m$  is smaller as is evident from fig. 3(b); the effect of surface energy on the fastest growing mode becomes increasingly less significant for large values of  $Y$ . Just as in (11), an analytic result can be derived for  $k_m$  for small values of  $\gamma/\mu h$ ,  $\nu \rightarrow 0.5$  and  $h(Y - Y_c)/\mu \ll 1$ :

$$hk_m(\nu, \frac{\gamma}{\mu h}) = hk_c(\nu, \frac{\gamma}{\mu h}) + \left( 0.39 \frac{\gamma}{\mu h} - 0.46(1 - 2\nu) \right) \frac{h}{\mu} (Y - Y_c) \quad (17)$$

Instability in a thin film whose surface experiences forces depends on three key sets of non-dimensional parameters namely the Poisson's ratio  $\nu$ , the normalised second derivative of the interaction potential  $hY/\mu$  and the normalised surface energy  $\gamma/\mu h$ . The whole picture of stability and bifurcation in this system and its dependence on the nondimensional parameters can be depicted pictorially as shown in fig. 4. Region *I* in fig. 4 is where the homogeneous solution is unique and stable while region marked *III* in the figure corresponds to the case when the homogeneous solution is “unphysical”, i.e., this analysis is not adequate. Region *II* is the most interesting – this corresponds to nearly incompressible material behaviour. In this region the homogeneous solution is unstable, with two possible elastic bifurcation modes; a viscoelastic analysis predicts a fastest growing mode with a wave vector that lies between the two elastic bifurcation modes.

We now turn to specific cases of the type of sys-

tem considered in this paper. First, we consider a rigid contactor interacting with the film via van der Waals forces. Assuming that the contactor is at a distance  $d$  above the undeformed surface of the film, the interaction potential  $U$  can be taken to be  $U(\mathbf{u} \cdot \mathbf{n}) = \frac{A}{12\pi(\mathbf{u} \cdot \mathbf{n} - d)^2}$  with  $F_0 = \frac{A}{6\pi d^3}$ ,  $Y = \frac{A}{2\pi d^4}$ . Taking the film to be made of rubber ( $\mu = 1$  MPa,  $\nu = 0.5$ ,  $\gamma = 0.1$  J/m<sup>2</sup>) and  $h = 1$  micron with  $A \approx 1$  eV. When  $d = 10$  nanometers we get  $hY/\mu = 1.6$  and for  $d = 5$  nanometers,  $hY/\mu = 25.6$ . Since the latter value is greater than  $hY_c/\mu$  which is 6.63 when  $\gamma/\mu h = 0.1$  (which is the present case), it is clear that the condition for bifurcation will be achieved as  $d$  is reduced from 10nm to 5nm. Thus as the contactor approaches the film, the film would buckle. This implies that the contact that forms between the contactor surface and the film will not be planar. We are not aware of any experimental work that can corroborate our results. We do, however, hope that the contents of this paper will be useful in designing experiments to verify our conclusions.

The second case considered is that of a film interacting with an external electric field. The system consists of two plates separated by a distance  $d$ ; the bottom plate is coated with a nearly incompressible polymeric film of height  $h$ . A potential difference of  $V$  is applied between the two plates. The quantity of interest is the value of the gap thickness  $d - h$  at which instability occurs in the film. The potential of interaction for this case is given by  $U(\mathbf{u} \cdot \mathbf{n}) = \frac{\epsilon_0 \epsilon_p V^2}{2(\epsilon_p d - (\epsilon_p - 1)(h + \mathbf{u} \cdot \mathbf{n}))}$  where  $\epsilon_0$  is the permittivity of free space,  $\epsilon_p$  is the dielectric constant of the polymer. Taking the mechanical properties of the polymer to be same as in the previous case, and taking  $\epsilon_p = 3$ , we get that the critical gap thickness  $d - h$  of 0.05 micron for a film of height 0.1 micron with the applied voltage of 100V. A gap thickness smaller than 0.05 micron will cause the film to buckle. It is evident that large electric fields are required to cause the instability.

We do wish to point out that this analysis is based on a linearised model, and will only provide the modes of instability, i. e., the wavelength of surface undulation and not the magnitude. A nonlinear analysis is required to obtain such a quantity and will be pursued in subsequent papers.

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- <sup>†</sup> vbshenoy@iitk.ac.in; Department of Mechanical Engineering
- <sup>‡</sup> ashutos@iitk.ac.in; Department of Chemical Engineering
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